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(54) Process for producing an isotropic separator for fuel cell

(57) The present invention provides a separator for a fuel cell in which an electrically anisotropic conductive filler can be made uniformly oriented without using a special mold or special molding apparatus, and which has excellent conductivity. This is achieved by (A) press moulding a conductive resin composition comprising an anisotropic conductive filler to obtain a plate-shaped product having front/back surfaces (10) and edge surfaces (20), (B) cutting said product in strip-shaped pieces (1b), (C) aligning said strip-shaped pieces such that the cut edge surfaces (20) form the back and front surfaces of the resulting plate and (D) press-forming the plate into its final shape.

FIG. 1 (A)

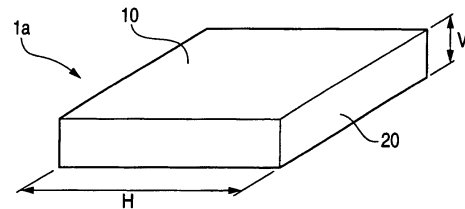


FIG. 1 (B)

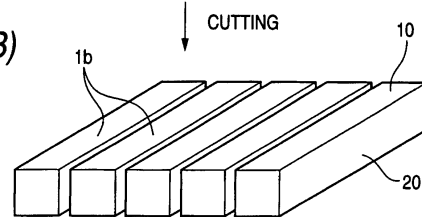


FIG. 1 (C)

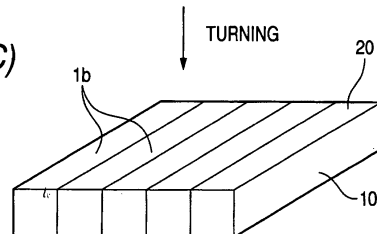
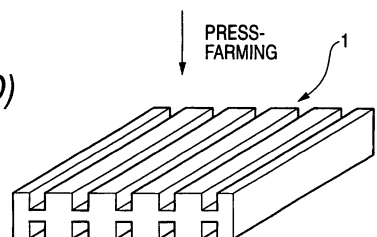


FIG. 1 (D)



**Description**

## FIELD OF THE INVENTION

5 **[0001]** The present invention relates to a process for producing a separator for fuel cells by molding a conductive resin composition comprising a resin and a conductive filler.

## BACKGROUND OF THE INVENTION

10 **[0002]** Recently, interest in fuel cells, in which the chemical energy possessed by a fuel is directly converted to electrical energy, is growing. In general, a fuel cell is in the form of a stack of many unit cells each comprising a matrix containing an electrolyte, electrode plates between which the matrix is sandwiched, and fuel-cell separators disposed on the outer sides of the electrode plates.

15 **[0003]** Fig. 3 is a perspective illustration showing an embodiment of a general fuel-cell separator 1. It comprises a flat plate part 11 and partition walls 12 rising on each side thereof at a predetermined interval. In fabricating a fuel cell, a large number of such fuel-cell separators 1 are stacked in the protruding direction of the partition walls 12 (upward/downward direction in the figure). As a result of this stacking, a pair of adjacent partition walls 12 form channels 13. A fuel passes on one side of each fuel-cell separator 1, while a gaseous oxidizing agent or the like passes on the other side. The fuel-cell separator 1 is hence required to have excellent gas impermeability so as to prevent the two ingredients from mixing with each other, and the partition walls 12 forming the channels 13 are required to have high dimensional accuracy.

20 **[0004]** On the other hand, the fuel-cell separator 1 is generally produced by compression-molding a conductive resin composition, prepared by kneading a conductive filler such as carbon black or graphite together with a resin such as an epoxy resin or phenolic resin, into a predetermined shape. However, there is a problem that since the conductive filler is not completely spherical, it is difficult to obtain a fuel-cell separator 1 in which the conductive filler is in a uniformly dispersed state. For example, since graphite is tabular, use thereof gives a fuel-cell separator 1 in which the graphite is apt to orient in layers in a horizontal direction in Fig. 3 (the direction indicated by the symbol H), resulting in poor conductivity in the thickness direction (the direction indicated by the symbol V).

25 **[0005]** For overcoming such a problem of conductive filler orientation, there is a technique in which the amount of a conductive filler to be incorporated is increased. However, the resultant conductive resin composition has reduced flowability and poor moldability, and this is apt to result in a decrease in dimensional accuracy. A technique that the composition is pressed from many directions during molding has also been proposed (see reference 1). This technique, however, has a disadvantage that it necessitates a special mold and a special molding apparatus and the molding step is complicated.

35 [Reference 1]

40 **[0006]** JP-A-8-180892 (the term "JP-A" as used herein means an "unexamined published Japanese patent application")

## SUMMARY OF THE INVENTION

45 **[0007]** An object of the present invention, which has been achieved in view of such a situation, is to provide a separator for fuel cells in which a conductive filler can be made uniformly oriented without using a special mold or special molding apparatus and which has excellent conductivity.

**[0008]** In order to achieve the above-mentioned objects, the present invention provides a process for producing a separator for fuel cells described below:

50 (1) A process for producing a separator for a fuel cell comprising the steps of: providing a conductive resin composition comprising a resin and an electrically anisotropic conductive filler; press-molding the conductive resin composition under pressure without heating to obtain a preformed product in the form of a flat plate; cutting the preformed product parallel to its edge face by a predetermined width to obtain strip-form preformed product pieces having a front surface, a back surface and a cut surface; aligning the strip-form preformed product pieces so as to form as a whole a plate shape having a front surface and a back surface each constituted by the cut surfaces of the strip-form preformed product pieces; and press-forming the whole preformed product pieces into a separator shape at a temperature not lower than a curing temperature of the resin.

(2) A process for producing a separator for a fuel cell comprising the steps of: providing a conductive resin composition comprising a resin and an electrically anisotropic conductive filler; press-molding the conductive resin

composition under pressure without heating to obtain a preformed product in the form of a flat plate; cutting the preformed product parallel to its edge face by a predetermined width to obtain strip-form preformed product pieces having a front surface, a back surface and a cut surface; aligning the strip-form preformed product pieces so as to form as a whole a plate shape having a front surface and a back surface each constituted by the cut surfaces and partially by the original front and back surfaces of the strip-form preformed product pieces; and press-forming the whole preformed product pieces into a separator shape at a temperature not lower than a curing temperature of the resin.

(3) The process for producing a separator for a fuel cell according to item 1, wherein the electrically anisotropic conductive filler is a conductive filler selected from the group consisting of: a tabular conductive material; a platy conductive material; or a fibrous conductive material.

(4) The process for producing a separator for a fuel cell according to item 3, wherein the electrically anisotropic conductive filler is expanded graphite.

(5) The process for producing a separator for a fuel cell according to item 1, wherein the conductive resin composition further comprising an electrically isotropic conductive filler.

## BRIEF DESCRIPTION OF THE DRAWINGS

### [0009]

Fig. 1 is process drawing showing steps of a process for fuel-cell separator production according to the present invention.

Fig. 2 is a schematic view showing the constitution of the measuring apparatus used in the Example of the present application.

Fig. 3 is a perspective illustration showing an embodiment of a separator for fuel cells.

## DETAILED DESCRIPTION OF THE INVENTION

**[0010]** The present invention will be explained below in detail.

**[0011]** Fig. 1 is process drawing showing steps of a process for fuel-cell separator production according to the present invention. First, a resin is sufficiently kneaded together with a conductive filler to prepare a conductive resin composition in the same manner as well-known method. When dry mixing is applied, it is suitable that the temperature in kneading process is controlled at lower than the softening temperature of the resin, preferably from 0 to 80°C. Further, the density of the mixture of the resin and conductive filler is preferably 40 to 300 kg/m<sup>3</sup>. On the other hand, when melt mixing is applied, it is suitable that the temperature in kneading process is controlled at higher than the softening temperature of the resin. The conductive filler may be suitably selected from ones that have hitherto been used for separators for fuel cells. In the present invention, an electrically anisotropic conductive filler is suitably used because the resin composition undergoes the steps which will be described later. The electrically anisotropic conductive filler indicates a conductive filler having different electronic resistances depending on the plane direction or axial of the conductive filler. As the electrically anisotropic conductive filler, a conductive material such as expanded graphite or a tabular, platy, or fibrous filler, is preferable in the present invention. In general, expanded graphite is obtained by treating flake graphite with concentrated sulfuric acid, and heating the treated graphite. The particular diameter, particularly the diameter of major axis, of the expanded graphite is preferably from 50 to 500 μm. The resin also is not particularly limited, and various thermoplastic resins and thermosetting resins can be used. The amount of the electrically anisotropic conductive filler to be incorporated also is not particularly limited. However, the amount thereof is preferably at least 40% by weight based on the whole conductive resin composition in view of securing the practical conductivity of the fuel-cell separator. On the other hand, the amount of the resin to be incorporated is preferably at least 20% by weight in view of the sealing properties and shape retention of the fuel-cell separator and flowability during molding. Further, in preparing the conductive resin composition, a conductive filler and a resin are preferably mixed at compounding ratio of from 40 to 80% by weight and from 20 to 60% by weight, respectively.

**[0012]** An electrically isotropic conductive filler may be added to the conductive resin composition according to need. The electrically isotropic conductive filler indicates a conductive filler having an uniform electric resistance on all of the plane direction or axial of the conductive filler. As the electrically isotropic conductive resin, a carbon black such as Ketjen Black, acetylene black, or furnace black is preferable in the present invention. These carbon blacks not only come into the spaces between layers of an oriented conductive material to form conduction paths and thereby function as a conductive filler, but also function as a reinforcement which enhances the mechanical strength of the fuel-cell separator to be obtained. Regarding the amount of the carbon black to be added, the ratio of the electrically anisotropic conductive filler to the carbon black is preferably from 1:1 to 1:4, in view of the effect of forming conduction paths, the reinforcing effect, and moldability.

**[0013]** Unless otherwise noted, the term "conductive filler" hereinafter stands for "electrically anisotropic conductive filler".

**[0014]** As shown in step (A) in Fig. 1, the conductive resin composition is packed into a mold having a cavity of a flat plate shape and pressed without applying heat thereto, thereby obtain a preformed product 1a in the form of a flat plate shape. The pressing conditions preferably include a pressure of from 50 to 100 MPa. In this preformed product 1a, the conductive filler is oriented in layers parallel to the flat part 10 (in the direction indicated by the symbol H) as an product provided by a well-known method.

**[0015]** Subsequently, as shown in step (B) in Fig. 1, the preformed product 1a is cut at a predetermined interval parallel to an edge face 20 to obtain preformed product pieces 1b in the form of strips. The width of the preformed product pieces is preferably from 0.1 to 20 mm.

**[0016]** Thereafter, as shown in step (C) in Fig. 1, the preformed product pieces 1b are turned over so that the cut surfaces (the surfaces parallel to the edge face 20) of each preformed product piece 1b face upward and downward, respectively, and then horizontally arranged so that the surfaces which originally constituted the flat part 10 come into face each other. As a result of this turning, the conductive filler in each preformed product piece 1b comes into the state of being oriented in layers in the original thickness direction (the direction indicated by the symbol V in step (A) in Fig. 1).

**[0017]** Thereafter, as shown in step (D) in Fig. 1, the resulting assemblage is placed in a mold for fuel-cell separators while maintaining the arranged state and pressed at a curing temperature or more for the resin in the same manner as well-known process, whereby a fuel-cell separator 1 is obtained. This forming with heating orients the conductive filler, which has been oriented in the direction indicated by the symbol V, again in the direction indicated by the symbol H, and disperses the conductive filler almost uniformly in each of the thickness direction and the plane direction as a whole. Thus, a separator 1 for fuel cells that has excellent conductivity is obtained. Namely, as demonstrated by the Examples set forth below, a separator for fuel cells that has a electric resistance reduced to about a half of that of separators produced through molding by conventional processes can be obtained.

**[0018]** According to the present invention, the conductive filler in the separator for fuel cells can be controlled so as to orient in any desired directions by changing the arrangement of the strip-form preformed product pieces 1 in step (C) described above. For example, when the strip-form preformed product pieces 1b are arranged without turning over some of the preformed product pieces 1b, then only that part of the resultant fuel-cell separator which corresponds to these preformed product pieces 1b without turning has lower conductivity than the other parts. Thus, there is a possibility according to the process of the present invention that an entirely new method of stacking fuel cells might be developed.

## EXAMPLES

**[0019]** The present invention will be explained below in more detail by reference to Examples and Comparative Examples, but the present invention should not be construed as being limited to these Examples.

**[0020]** The conductive materials and resins shown below were mixed at compounding ratio (wt. %) shown in Table 1 to produce samples by the process set forth below.

### Conductive Materials

**[0021]** Expanded graphite  
(particle diameter; about 400-800  $\mu\text{m}$ )

Carbon black  
(acetylene black; particle diameter; about 5-10  $\mu\text{m}$ )

**[0022]** The particle diameter of the expanded graphite described above stands for the diameter of major axis. The particle diameter of the carbon black stands for the primary particle diameter.

### Resins

**[0023]**

Thermosetting epoxy resin

(Epoxy resin F6136, manufactured by SOMAR CORP., softening temperature is 80 °C)

Thermoplastic polymethylpentene (PMP)

(Polymethylpentene RT18, manufactured by MITSUI CHEMICALS, melting temperature is 240 °C)

Thermoplastic liquid-crystal polymer (LCP)

(Rodrun LC5000F, manufactured by UNITIKA LTD., melting temperature is 280 °C)

Production Process

5 [0024] The ingredients were kneaded by dry mixing in Example 1 or by melt mixing in Examples 2 and 3 in Table 1 to obtain a kneaded mixture in each Example. For details, when dry mixing was applied, predetermined amounts of each ingredient were set in a kneading space of 10L Henschel mixer, then the ingredients were mixed in the mixer for 120 seconds at room temperature. On the other hand, when melt mixing was applied, the resins were set in the kneading space and melted by heating in Brabender (or Labo-plastomill), then an electrically anisotropic conductive filler and an electrically isotropic conductive filler were added thereto and kneaded with the resins in this order, for 15 minutes, at 260 °C when polymethylpentene is used, at 300 °C when liquid-crystal polymer is used. The kneaded mixture obtained in Example 1 was molded into a preformed product in the form of a plate shape at ordinary temperature and a pressure of 20 MPa. Subsequently, the preformed product of Example 1 was cut into strips at an interval of 5 mm. These strips were set in a mold so that the cut surfaces became upper and lower sides, and pressed at 180°C and a pressure of 100 MPa to obtain a test piece for Example 1 in the form of a 100 x 100 x 5 mm sheet-form. On the other hand, test pieces for Examples 2 and 3 were obtained from the kneaded mixtures of Examples 2 and 3 in the same manner as in Example 1, except that the pressure in preformed formation was changed to 100 MPa. Further, on the other hand, with respect to each of Comparative Examples 1 to 3, kneaded products were prepared in the same manner as Example 1 to 3, respectively. Then the kneaded products were pressed and molded into the form of a sheet at 180 °C and a pressure of 100 MPa to obtain test pieces for Comparative Examples 1 to 3, respectively.

Measurement of Resistance

20 [0025] Resistances of the test pieces produced by the above process were examined by using the measuring apparatus shown in Fig. 2. The measuring apparatus shown in the figure has the following constitution. Namely, a test piece 30 is sandwiched between a pair of gold-plated electrodes 50 via a carbon paper 40 on each side. A constant current 1 A is caused to flow from a power source 10, and the voltage between the carbon papers 40 is measured with a voltmeter 60. Then resistances is calculated from the constant current and the measured voltages by using the equation  $V=IR$ . The calculated resistance was multiplied by the surface of the sample to obtain the total resistance. Accordingly, it is noted that each total resistance includes the contact resistance between each carbon paper 40 and the test piece 30 and the volume resistance of the test piece 30. The results of the measurement are given in Table 1.

Table 1

	Example 1	Comparative Example 1	Example 2	Comparative Example 2	Example 3	Comparative Example 3
Epoxy resin	30	30	-	-	-	-
PMP	-	-	40	40	-	-
LCP	-	-	-	-	50	50
Expanded graphite	60	60	50	50	40	40
Carbon black	10	10	10	10	10	10
Process of Production	A → B	B	A → B	B	A → B	B
Resistance (mΩ·cm <sup>2</sup> )	20	40	30	50	50	80

Note 1) The amount of each ingredient is given in % by weight.

Note 2) In "Process of production", "A" indicates orientation regulation and "B" indicates compression molding.

[0026] The test pieces of Comparative Examples 1 to 3 shown in Table 1 are those provided by the conventional process. The results show that the test pieces of Examples 1 to 3, which were molded according to the present invention, have a resistance reduced to about a half of the resistance for the test pieces of the Comparative Examples.

[0027] As described above, according to the present invention, the orientation of a conductive filler can be made uniform and a separator for fuel cells which has excellent conductivity is obtained without using a complicated mold.

[0028] While the present invention has been described in detail and with reference to specific embodiments thereof,

it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing the spirit and scope thereof.

[0029] The present application is based on Japanese Patent Application No. 2003-091810 filed on March 28, 2003, and the contents thereof are incorporated herein by reference.

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## Claims

1. A process for producing a separator for a fuel cell comprising the steps of:

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providing a conductive resin composition comprising a resin and an electrically anisotropic conductive filler; press-molding the conductive resin composition under pressure without heating to obtain a preformed product in the form of a flat plate;

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cutting the preformed product parallel to its edge face by a predetermined width to obtain strip-form preformed product pieces having a front surface, a back surface and a cut surface;

aligning the strip-form preformed product pieces so as to form as a whole a plate shape having a front surface and a back surface each constituted by the cut surfaces of the strip-form preformed product pieces; and press-forming the whole preformed product pieces into a separator shape at a temperature not lower than a curing temperature of the resin.

20

2. A process for producing a separator for a fuel cell comprising the steps of:

25

providing a conductive resin composition comprising a resin and an electrically anisotropic conductive filler; press-molding the conductive resin composition under pressure without heating to obtain a preformed product in the form of a flat plate;

cutting the preformed product parallel to its edge face by a predetermined width to obtain strip-form preformed product pieces having a front surface, a back surface and a cut surface;

30

aligning the strip-form preformed product pieces so as to form as a whole a plate shape having a front surface and a back surface each constituted by the cut surfaces and partially by the original front and back surfaces of the strip-form preformed product pieces; and

press-forming the whole preformed product pieces into a separator shape at a temperature not lower than a curing temperature of the resin.

35

3. The process for producing a separator for a fuel cell according to claim 1,

wherein the electrically anisotropic conductive filler is a conductive filler selected from the group consisting of: a tabular conductive material; a platy conductive material; or a fibrous conductive material.

40

4. The process for producing a separator for a fuel cell according to claim 3,

wherein the electrically anisotropic conductive filler is expanded graphite.

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5. The process for producing a separator for a fuel cell according to claim 1,

wherein the conductive resin composition further comprising an electrically isotropic conductive filler.

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FIG. 1 (A)

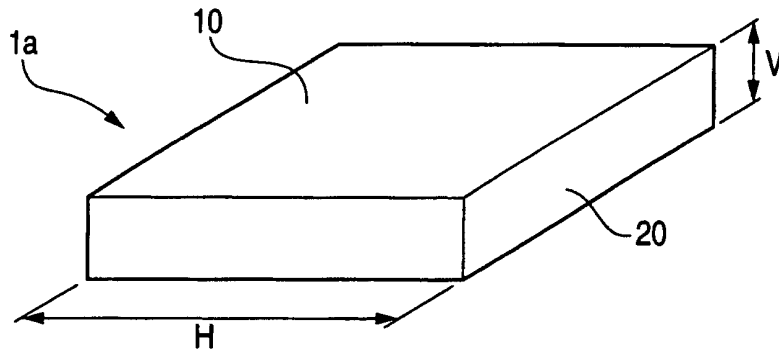


FIG. 1 (B)

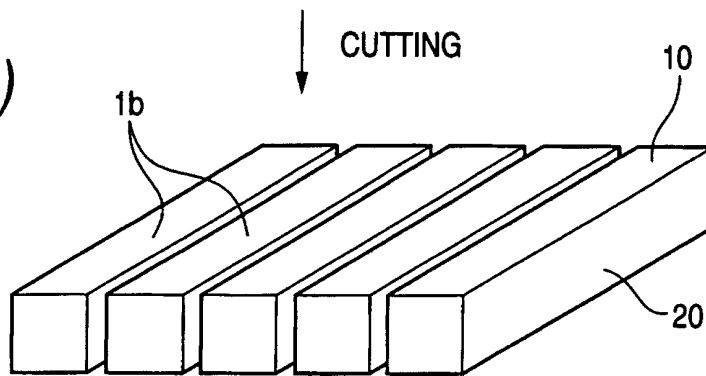


FIG. 1 (C)

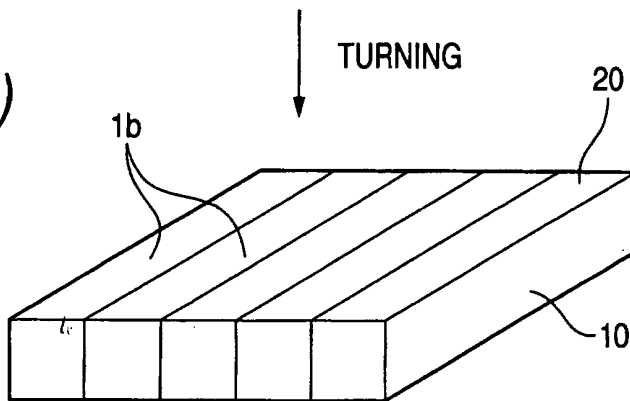


FIG. 1 (D)

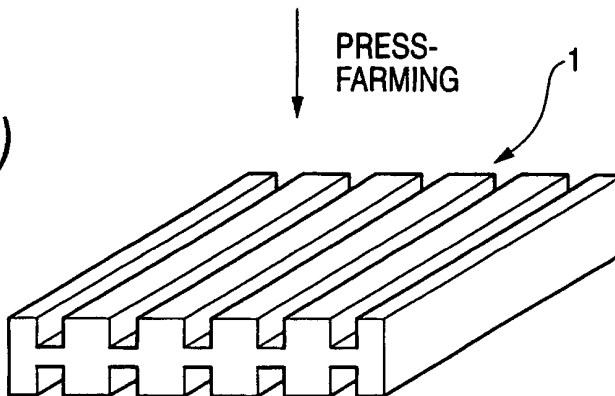


FIG. 2

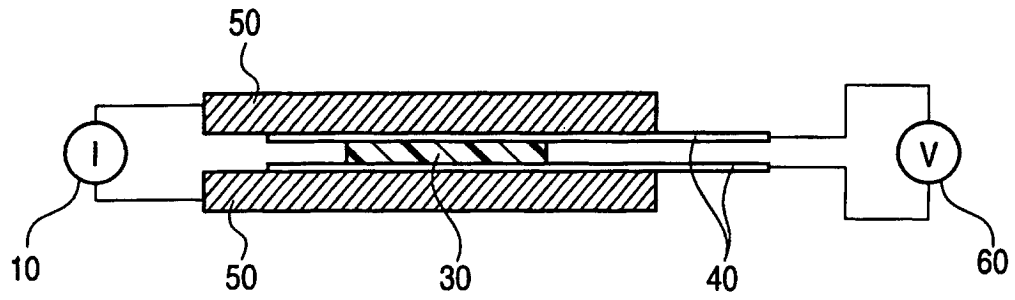
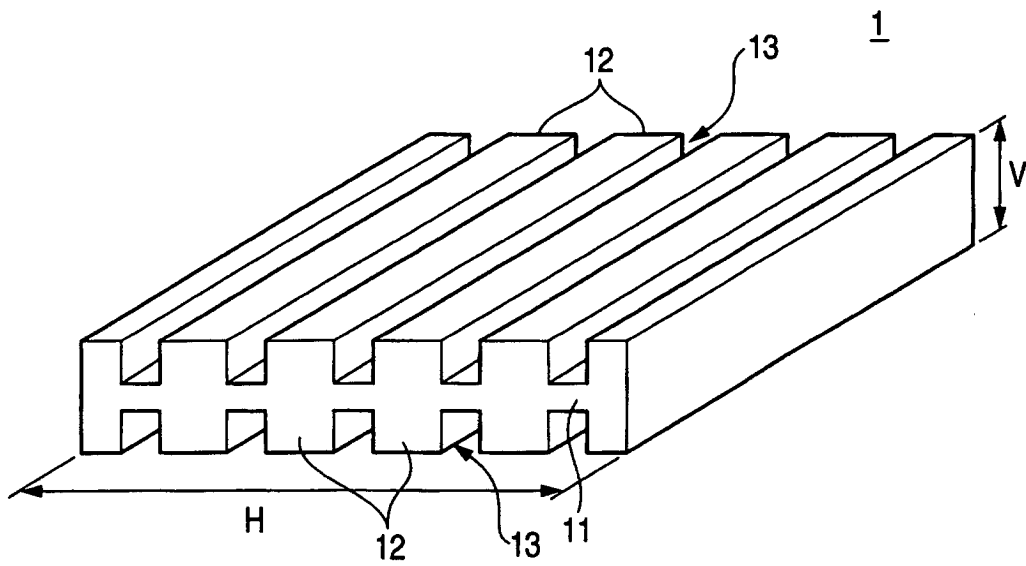


FIG. 3





DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
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A	----- EP 1 213 272 A (SGL CARBON AG) 12 June 2002 (2002-06-12) * paragraphs [0031], [0032], [0044] * * examples 4,5 *	1-5	TECHNICAL FIELDS SEARCHED (Int.Cl.7) H01M
The present search report has been drawn up for all claims			
Place of search Munich		Date of completion of the search 8 July 2004	Examiner Jacquinot, P
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document	

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ANNEX TO THE EUROPEAN SEARCH REPORT  
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